CHARACTERIZATION OF THE BRICKS USED IN THE CONSTRUCTION OF THE ALCAZABA (MÁLAGA, SPAIN)

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SUMMARY 
The Alcazaba of Málaga is mainly built with manufactured materials (bricks, mortars and rammed-earth walls). It was constructed and reconstructed from VIIIth to XVth Centuries. The state of conservation of its structure on some zones is relatively bad, which makes necessary a study to determine the causes and mechanisms of weathering. Besides, the constructive chronology on several areas is very uncertain, for what a relative datation study is being carried out with base on the mineralogical and petrographical characterization of bricks of known chronology, belonging to the most important building periods (XIIth and XIIith-XIVth centuries). Besides, this study allows to determine the source of the raw materials of the bricks. 

Keywords: Alcazaba, bricks, mortars, vitrification, datation, conservation.

1. INTRODUCTION
The Alcazaba is one of the most representative historical buildings of Málaga. It is considered to be one of the most symbolic of the military fortifications from the Islamic period in Spain and was built and rebuilt continuously, but most particularly from the VIIIth to the XVth centuries.
The poor state of conservation of certain areas needing to be restored, together with the limited information available on the exact chronology of the erection of particular zones, have led to an extensive research project aimed at characterizing the materials in the monument.
Since the Alcazaba was primarily constructed with manufactured materials (bricks, mortars, rammed-earth walls), the description of these types of materials in the different areas for which there is a degree of certainty regarding the chronology could help to establish the age of materials in zones of uncertain date in cases where there are noticeable differences between periods. In addition, this research will provide knowledge on the origin of primary materials and the technological level employed in their manufacture.
Another objective of this project was to determine the overall state of conservation of the building, together with the causes and weathering processes that have influenced its state, thus allowing corrective measures to be proposed in the conservation-restoration project to be undertaken.
Included within this paper is a preliminary study carried out mainly on brick samples taken from what seem to be the periods of greatest building activity in the Alcazaba (the XIth, and the XIIIth to the XIVth centuries). In general, the conservation state of the bricks is good in spite of large amounts of gypsum detected in a great number of them. In this case, the gypsum does not appear to be responsible for significant disruption, rather, the alteration seems to be more related to the quality of the bricks, being dependent on the primary material as well as on the baking temperatures reached.

2. MATERIALS AND METHODS.
We have taken samples from various zones of the Alcazaba showing archaeological evidence allowing them to be attributed to particular periods (the XIth, and the XIIIth to XIVth centuries). The object of these samplings is to establish affinities between materials belonging to the same period of construction, which could aid in establishing relative dating in those areas of unknown age.
In total we took nine brick samples, six of which are from the XIth century (Puerta de las Bóvedas: PB1, PB2; Puerta de las Columnas: PC1, PC2; Torre de los Abencerrajes: TA1; Torre del Homenaje: TH1) while
the other three are from the XIIIth to the XIVth centuries (Puerta del Cristo: PX1, PX2; Torre de los Abencerrajes: TA2).

At the same time, samples were taken from the mortar joints of each brick to see whether there was any relationship between the composition of the mortar and the secondary phases found in the bricks. This analysis was undertaken due to the finding in previous studies of very high gypsum contents in some bricks in the Alcazaba.

Techniques used for the characterization included X-ray diffraction (XRD) for the determination of the major minerals in the total sample of both bricks and mortars; polarized light microscopy using thin sections to study the overall texture of the bricks, the nature of the temper, the vitrification state of the matrix, the secondary mineral phases, and the origin of the primary materials; scanning electron microscopy (SEM) including EDX to determine the degree of vitrification in each brick and to identify minerals and weathering products undetectable by other means.

3. RESULTS.

3.1. X Ray Diffraction.

The XRD results for the bricks are summarised in Table 1, where it may be seen that the bulk mineral composition is expressed as a % of the samples, with the chronology indicated for each one.

The phyllosilicate heading, it should be noted, includes both clay minerals comprising part of the matrix as well as the phyllosilicates (micas) that may form part of the temper, which is made of fragments of metamorphic rocks (phyllices and slates) identified by light microscopy.

As may be seen, there are no clearly marked differences between the bricks from different periods. Taking all the samples from the XIIIth century (except PC1 and TA1, which are of a different nature, as indicated by the petrographic analysis), abundance in the mineralogical association is very similar. High-temperature phases (D-W, Gh) are common in these bricks, while phyllosilicates are quite scarce, occurring in inverse proportion to calcium silicate contents.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Q</th>
<th>Fd</th>
<th>Ph</th>
<th>D-W</th>
<th>Gh</th>
<th>Gp</th>
<th>Cc</th>
<th>Ox</th>
<th>Age</th>
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<td>15</td>
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</tr>
<tr>
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<td>20</td>
<td>8</td>
<td>10</td>
<td>8</td>
<td>10</td>
<td>5</td>
<td>15</td>
<td>XI</td>
</tr>
<tr>
<td>PB2</td>
<td>30</td>
<td>20</td>
<td>15</td>
<td>10</td>
<td>5</td>
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<td>&lt;5</td>
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<td>XI</td>
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<tr>
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<td>34</td>
<td>&lt;5</td>
<td>45</td>
<td>&lt;5</td>
<td>14</td>
<td>5</td>
<td>XI</td>
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<td></td>
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<td>9</td>
<td>10</td>
<td>8</td>
<td>5</td>
<td>13</td>
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<td>8</td>
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<tr>
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<td>10</td>
<td>&lt;5</td>
<td>&lt;5</td>
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<tr>
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<td>12</td>
<td>26</td>
<td>&lt;5</td>
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<td>18</td>
<td>7</td>
<td>XIII-XIV</td>
<td></td>
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<tr>
<td>TH1</td>
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<td>7</td>
<td>&lt;5</td>
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</table>

Q: quartz; Fd: feldspars Ph: phyllosilicates; D-W: diopside-wollastonite; Gh: gehlenite; Gp: gypsum; Cc: calcite; Ox: oxides (hematite+magnetite).

It should be pointed out that in sample PB1 the inside and outside of the brick were analysed separately since it was observed in the hand sample that there was a colour change from orangeish ochre on the inside to a more intense red on the outside. As noted in the table, there are no
noteworthy differences with regard to the proportion of high-temperature phases, which indicates that the baking time was long enough for the mineralogy to be uniform, indicating that the colour change must be due to other causes.

At least two of the three bricks from the Xlth to the XIVth centuries (PX2 and TA2) have a mineralogy clearly indicative of lower baking temperatures - high phyllosilicate contents and very low high-temperature phase contents. Sample PX1 is very similar in composition to the bricks from the XIth century, which had longer baking times.

In addition, and independently of the age, there seems to be quite a direct relationship between the feldspar content and the baking temperature, as the feldspars increase concomitantly with the content in high-temperature phases. This circumstance indicates that at least part of the feldspar content could correspond to calcium feldspars formed during the baking process.

The same tendency seems to exist for the iron oxides, the high content of which is also responsible for the reddish colour of these bricks.

Table 2 presents the XRD results for the samples of joint mortars for some of the bricks.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Q</th>
<th>Fd</th>
<th>Ph</th>
<th>Gp</th>
<th>Cc</th>
<th>Dta</th>
<th>Age</th>
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<td>7</td>
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<td>14</td>
<td>XI</td>
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<td>PB2M</td>
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<td>&lt;5</td>
<td>10</td>
<td>62</td>
<td>&lt;5</td>
<td>20</td>
<td>XI</td>
</tr>
<tr>
<td>PC2M</td>
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<td>5</td>
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<td>&lt;5</td>
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<td></td>
<td>XI</td>
</tr>
<tr>
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<td>15</td>
<td>&lt;5</td>
<td>90</td>
<td>XIII-XIV</td>
<td></td>
</tr>
<tr>
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<td>7</td>
<td>72</td>
<td>XIII-XIV</td>
<td></td>
</tr>
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<td>8</td>
<td>44</td>
<td></td>
<td></td>
<td>XIII-XIV</td>
</tr>
<tr>
<td>TH1M</td>
<td>47</td>
<td>&lt;5</td>
<td>8</td>
<td></td>
<td>44</td>
<td></td>
<td>XI</td>
</tr>
</tbody>
</table>

The mineralogical composition of the mortars is quite varied, as may be seen, ranging from gypsum-rich mortars to lime mortars (either dolomitic lime or calcitic).

In broad terms, a comparison of the results reveals the compositional similarity that can be established based more on chronology than on location in the Alcazaba. We will not enter here into details since focuses on the composition of the mortars and their effect on adjoining bricks. In this respect, the most notable characteristic is that there is no apparent relationship between the gypsum content of some bricks and the composition of the surrounding mortar, although in some cases there does seem to a connection (eg. samples PB1 and PB2).

In contrast, it is possible to establish a relationship between the increase in calcite of bricks in contact with calcitic lime mortars, indicating that the overall calcite content in these bricks may have risen due to dissolution of part of the mortar and subsequent recrystalisation inside the bricks.

3.2. Optical Microscopy.

Thin sections of all the brick samples were prepared for petmographic study. The samples from the XIth century (PB1, PB2, PC2, and TH1) and sample PX1 from the XIIth to the XIVth centuries all have very similar petmographic characteristics. In plane-polarized light all of them have a dark-reddish, almost black, matrix that in most cases points to a high degree of vitrification. In some bricks, examination under crossed polar revealed areas with a microcrystalline matrix in which certain minerals can be identified, particularly phyllosilicates.

The temper of these bricks is basically comprised of quartz and fragments of metamorphic rocks (phyllites and slates) that are very rich in phyllosilicates showing clear reaction signs. In several cases feldspars and
fragments of previous pottery "chamotte" have also been identified. The pottery remains stand out clearly from the rest of the paste since they have a more vitreous matrix and a finer grain size. The size of the temper fragments varies from brick to brick. In samples PB1 and PB2, the quartz grains range widely in size, from 2mm ("chamotta") and quartz fragments that can be observed in hand samples) to 0.2mm, with shapes ranging from subrounded to subangular. In the rest of the bricks from this group, the temper fragmentes are smaller and more homogeneous. These bricks also preserve the most birefringent zones from the original paste texture.

Within this group, samples with greater vitrification have darker and more vitreous nodular zones that can be clearly differentiated from the rest of the matrix due to retractive cracking. These areas could be zones of a different composition, the result of uneven mixing of the original paste, with the cracking occurring during cooling (Figure 1).

Porosity is quite high in the bricks from this group, but light microscopy shows it to consist of very rounded macropores, retractive cracking, and fissures.

The other bricks (PC1, TA1 - Xth century; and PX2, TX2 - XIIth to XIVth centuries) are texturally very different from those described above. A poorly vitrified matrix is common in all of them, particularly in sample PC1 where there are practically no indications of vitrification. Examination in plane-polarised light reveal the matrix to be orangeish brown, while crossed polars show it to be very birefringent and phyllosilicate-rich (Figure 2). In all the samples except PC1 there are dark-red areas that are nearly black and of lesser birefringence, indicating greater vitrification. These areas are more common towards the edges of the bricks and more extensive the higher the baking temperature.

The temper of these bricks is similar to that of the afore-described bricks, with only the size and homogeneity of the temper fragments varying between the two groups. The most noteworthy difference is that in sample TA1 the size of both the quartz and the rock fragments is much larger and more homogeneous, while at the same time the quartz is quite rounded (perhaps having been added by the brickmaker).

In sample PC1, the temper, aside from containing the previously described additions, also has abundant foraminifer remains (nummulites and globigerinas). This is the only brick in which these microfossils have been identified (Figure 3). Another distinguishing characteristic of this brick is the presence of calcite crystals clearly forming part of the temper. These two factors indicate that the primary material used in its manufacture was likely different.

The porosity of all these bricks is also quite high, although the pores are not as large as in the previous set of bricks. In addition, there are some cases of superficial fissures parallel to the surface and to the orientation of the matrix phyllosilicates.

All of the bricks, indepently of the age, contain gypsum in varying amounts, preferentially filling pores and fissures (Figure 4), but in some cases it has also been identified, by its low birefringence, comprising part of the matrix.

Calcite that is clearly secondary has been found in several bricks filling some pores and fissures. In other cases concentrations of micritic calcite have been observed, of unknown origin.


SEM has been used to examine the fracture surfaces in samples PB1, PX1, PA2, and TH1 in order to ascertain the degree of paste sintering achieved in each brick.

The samples from the Xth century (PB1 and TH1) are highly vitrified (Figures 5 and 6), although in neither is it extensive, as proven by the laminae of phyllosilicates around the vitrified zones. Nevertheless, in the samples from the XIIIth to the XIVth centuries there are scarce or no signs of vitrification (figure 7), with the matrix the same as the original paste must have been.

EDAX microanalysis has revealed the presence of certain accessory minerals, mostly heavy, in the matrix, such as pyrite, zircon, and even monazite. Soluble salts were also found in abundance (ClNa: halite). This salt (Figure 8) appears distributed across the surface of all the samples and may directly affect the deterioration of the bricks.
4. CONCLUSIONS.

The raw material used for all the bricks analysed was the same and contained large amounts of fragments of metamorphic rocks (phyllites or slates). In some cases, such as in sample PC1, the presence of foraminiferae would seem to indicate either another source area or deliberate mixing by the brickmaker for unknown reasons.

The fact that gypsum has been found in all the bricks, and its presence in their matrix, albeit in small amounts, means it cannot be attributed exclusively to adjoining mortars since in many cases the mortar is of lime. This situation leads us to conclude that at least in part the gypsum likely originates from the primary material.

On examination of the geological material in the areas surrounding Málaga, it has been ascertained that the mineralogical composition of these bricks could coincide with the Red Facies clays from the Upper Complex of the Maláguid Cover. These facies crop out quite close to the Alcazaba and contain white gypsum, often intercalated with gypsum containing greater amounts of clay. Iron-rich red sandstones have been described in association with these clays, as well as frequent heavy minerals such as zircon and pyrite. Information is lacking to attribute the source of the bricks to a specific area.

The temper of the bricks consisting mainly of quartz and rock fragments seems to correspond to the original material (natural temper), except for the fragments of crushed pottery (chamotte), which must have been added.

The carbonate content is low in nearly all the samples, almost always appearing as secondary calcite, with relic-type calcite being in general quite scarce. The original material must therefore not have been very rich in calcite, since even less-baked bricks, in which the original calcite should have been at least in part preserved, have low concentrations of calcite. Nevertheless, the calcite must have existed in high enough amounts to react with the clays and transform itself into high-temperature calcium phases in those bricks with abundant amounts of such phases. This may be one of the reasons why the sintering reached by the paste was lesser, since the flux influence of the calcium on the bricks may have been slight.

The baking temperatures must have reached at least 800-850°C in all the bricks, since they all contain greater or lesser amounts of high-temperature phases (except sample PC1). In the XIth century bricks with high calcium silicate contents, this temperature may have been greater or the baking time longer, as the matrix is vitreous and the phyllosilicates that appear derive almost entirely from fragments of metamorphic rocks, which would indicate that, although the baking temperature may have been higher for these bricks, it was still not high enough to completely decompose these fragments.

Generally speaking, the XIth century bricks (except samples PC1 and TA1, which are distinct in composition and texture) were baked at higher temperatures and consequently are more vitrified. In addition, they are texturally and compositionally very similar. In contrast, the samples from the XIIIth to the XVth centuries were baked at lower temperatures and have scarce or no vitrification. The only exception is sample PX1, which dates from this period but is more similar to the XIth century bricks, admitting the possibility that this brick was indeed manufactured in the XIth century and subsequently reused.

In order to confirm these hypotheses, research will continue with further samples to obtain greater statistical reliability. If the results of these studies are positive we will have a tremendous archeometric reference extrapolable to other zones of the Alcazaba.

The presence of concentrations of CaNa that were detected by SEM could originate from marine aerosols, since the Alcazaba is located on the coast, where the action of such aerosols may have caused this salt to precipitate inside the bricks.

The state of conservation of the bricks is fairly good considering their high content in soluble salts (gypsum and halite). This fact may perhaps be explained by the great presence of macropores and low amounts of micropores, which latter are the most at risk from the decrystallisation of salts.
Figure 1. Nodular zones in XI century's bricks.

Figure 2. Global texture of XIII-XIV century's bricks.

Figure 3. Brick with Foraminifera (PC1)

Figure 4. Gypsum in bricks cracks.

Figure 5 and 6. Vitrification in XI century samples

Figure 7. No vitrification in XIII-XIV century's samples.

Figure 8. Halite crystals in bricks.
REFERENCES


